Bridging the Length and Time Scales from Atomistics to the Microscale

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Polymer networks of interest to LLNL's mission undergo structural and mechanical changes over time. These changes occur due to radiation damage, contaminant transport and reaction, and prolonged deformation due to physical loads. To date, useful molecular dynamics (MD) and finite element analysis (FEA) efforts have been performed. These efforts, however, focus on the nano and continuum scales, respectively. The missing link in this effort is a "meso" or micron-to-centimeter scale that uses interaction potentials generated by MD simulations and supplies time-dependent material properties to continuum models. This last year has resulted in a capability to generate 3-D foam networks with the ability to track Brownian walkers with steric effects and local interaction potentials derived from MD simulations.

Project Goals

The goal of this project is to bridge the length and time scales from atomistics to microscale.

Relevance to LLNL Mission

LLNL has an interest in polymer networks and their structural changes. A fundamental numerical approach to understand and characterize what happens to the micro and nanostructures of polymeric materials is a central need of LLNL's commitment to DOE's Enhanced Surveillance Campaign.

FY2007 Accomplishments and Results

The mechanical properties of polymeric materials are a function of the bonds between crosslinks and the crosslink density of the material. Given that the bulk material length scales are typically on the order of centimeters and

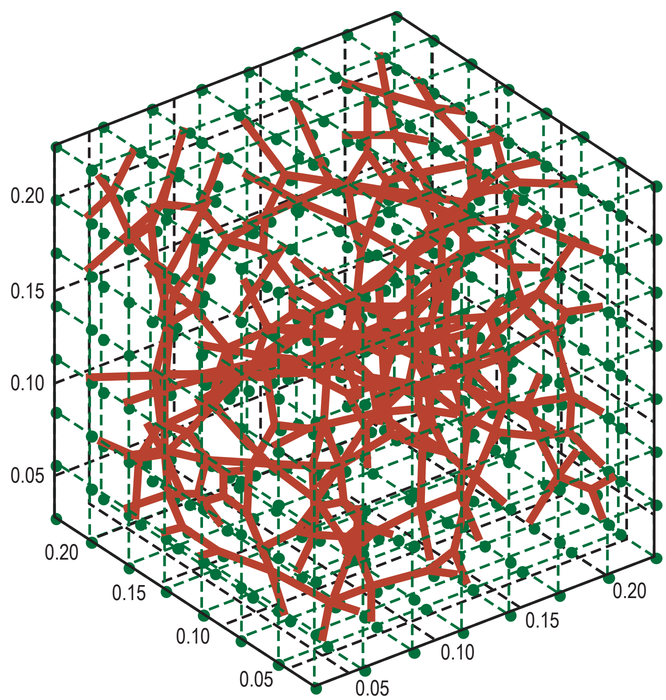

Figure 1. 3-D vertex model of a polymer foam. The final structure (shown in red) is evolved from an initial cubic lattice (shown in green).

the polymer structure is on the order of nanometers, modeling of polymeric materials is inherently a multiscale problem. In the work being performed on this project, a 3-D representation of the microstructure with a quantifiable crosslink density was adapted from a vertex model. This model allows the user to prescribe mechanical properties to the bonds between crosslinks, such as Hookean springs, and allows automatic tracking of crosslink density. Additionally, the numeric polymeric media can be mechanically assayed, using tension, compression and shear, to yield continuum property information. In parallel with this adaptation of the vertex model, a Brownian dynamics simulation was adapted to track random walkers through the 3-D medium.

These random walkers "react" with the medium at crosslinks in accord with the interaction potentials generated by MD simulation. It has been shown that the modulus of polymeric material is directly dependent on the crosslink density of the material. As the random walkers traverse the medium as a function of time, they sever crosslinks, modifying the modulus of the material and yielding a time-dependent change in mechanical properties.

This approach can also be modified to track the interaction of radiation with the polymeric medium while accounting for the degradation in bonds, crosslinks, and resulting mechanical properties. Furthermore, the vertex model provides a starting point to get a glimpse of polymeric microstructure of real systems. With some calibration to experimental data, this model can be used to postulate microstructural properties.

To date, we have implemented a 3-D vertex polymer foam model that enables a new way to assess bulk material properties (Fig. 1). The model also includes a capability to characterize contaminant diffusion in the foam (Fig. 2). Our programmatic customers see promise in this mesoscale approach since it is less costly than MD simulations and they would like to further enhance the current model.

Related References

1. Grimson, M. J., "Structure and Elasticity of Model Gel Networks," *Mol. Phys.*, **74**, pp. 1097-1114, 1991.
2. Flory, P. J., Principles of Polymer Chemistry, Cornell University Press, Ithaca, New York, 1953.

FY2008 Proposed Work

We have submitted a proposal to incorporate these enhancements and validate with experiments. Continuing the work would have high impact to the program since it aligns with their goals and would also strengthen the relationship between Engineering and the Enhanced Surveillance Campaign.

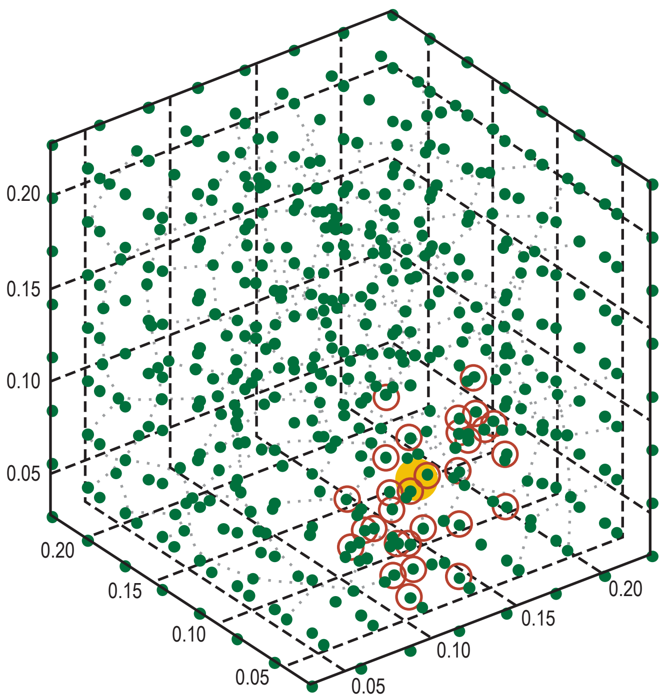

Figure 2. Contaminant particle (gold) represented by a random walker in the vertex model. Crosslinks within the containment's range of influence are highlighted in red.